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# VISCOSITY MEASUREMENTS OF DILUTE SOLUTIONS OF HELIUM-3 IN SUPERFLUID HELIUM-4 BETWEEN 0.1 AND 1.2 K

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16. Abstract

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## SUMMARY

Viscosity measurements of three dilute solutions of helium-3 ($He^3$) in superfluid helium-4 ($He^4$) were made between 0.1 and 1.2 K for concentrations of 1.09, 1.57, and 4.67 atoms of $He^3$ per 100 atoms of solution. A torsional crystal technique with a particularly low rim velocity was employed. As the temperature decreased, a slight change in slope became noticeable near the Fermi temperatures $T_f$ of each solution. The experimental results were in qualitative agreement with the theoretical predictions of Baym and Saam, and, near 1.0 K, with the only previously available experimental values. However, in the case of the 1.09- and 1.57-percent solutions, the values were a factor of 2 or more below the theoretical ones. In addition, for these solutions, a sharp change in slope occurred near 0.3 K (which is not near $T_f$) as compared with the very slight and gradual drop in the theoretical values. If the interaction between the $He^3$ quasi-particles and the $He^4$ phonons had been larger than the estimate used to make the theoretical calculations, the agreement with the experimental results would have been improved.

## INTRODUCTION

The possibility that helium-3 ($He^3$) could actually dissolve in helium-4 ($He^4$) and form a single phase that persists down to $T = 0$ K was mentioned in an early paper of Landau and Pomeranchuk (ref. 1), which considered the effects of impurities in liquid $He^4$. Experimental indication that a single phase did, in fact, exist for dilute mole fractions of $He^3$ at absolute zero was provided by Edwards et al. (ref. 2) in 1965. These investigators measured the specific heat of dilute solutions of $He^3$ in $He^4$ and concluded that $He^3$-$He^4$ solutions having mole percents of 6 or less of $He^3$ form a stable single phase down to absolute zero. (For future reference, 1 mole percent is defined as one atom of $He^3$ per 100 atoms of solution.)

$He^3$-$He^4$ mixtures (whether or not they form a single phase) have been of great theoretical interest because they are realizations of a dilute Fermi system. A number of measurements and theoretical calculations of various properties of these mixtures showed that the effective $He^3$-$He^3$ interactions are quite weak in dilute solutions, whereas, in pure $He^3$, these interactions are strong. Bardeen, Baym, and Pines (ref. 3) constructed an interaction potential for dilute solutions that enabled semiquantitative estimates of some quantities to be made which were in reasonable agreement with experiment. Refinements of this theory subsequently enabled rather detailed computations to be made of the transport properties of these dilute solutions, such as thermal conductivity and viscosity, as a function of temperature (refs. 4 and 5). In particular, Baym and Saam (ref. 6) computed the viscosity for two $He^3$-$He^4$ solutions having mole percents of 1.3 and 5.0, respectively. These computations were made using the assumption that, at low temperatures ($T < 0.6$ K), the transport coefficients will be largely determined by the $He^3$. This assumption seems reasonable since transport by the elementary excitations in $He^4$ is, in fact, limited by their interaction with the $He^3$.

The predicted behavior of viscosity $\eta$ for decreasing temperature included a minimum (see figs. 4 and 5). Since the only available data for the viscosity of such mixtures were for temperatures above 1 K (ref. 7), no experimental check on this behavior could be made. For this reason, this investigation to measure the effective viscosity of solutions of $He^3$ in superfluid $He^4$ as a function of temperature below 1.2 K is very timely. The three molar concentrations of $He^3$ used herein were 1.09, 1.57, and 4.67 percent. The temperature range covered was 0.1 to 1.2 K, so that these measurements should provide a reasonable test of the theoretical predictions.

There may be some question as to whether or not the quantity measured and herein called the "effective viscosity $\eta_E$" is the true viscosity. The measurements were made using a torsional crystal technique described previously (ref. 8). Herein, the same relation is used between the viscosity and the logarithmic decrement of the crystal as that used previously for pure liquid $He^3$ and $He^4$ (ref. 8):

$$\Delta_l^2 = \frac{S^2 \eta_E \rho_{NS}}{FM^2} \tag{1}$$

where $\Delta_l$ is the crystal decrement due to the liquid, S is the total crystal surface area, F is the resonant frequency, and M is the crystal mass. Finally, an effective normal density of solution $\rho_{NS}$ was used for the various mixtures given by (ref. 9):

$$\rho_{NS} = \rho_{no} + \rho_S \frac{m_3^*}{m_4} X \qquad \text{for } T < 1.2 \text{ K}$$

where $\rho_{no}$ is the normal density of pure $He^4$, $\rho_S$ is the actual density of the solution, $m_3^*$ is the effective mass of a $He^3$ atom (2.34 $m_3$), $m_3$ and $m_4$ are the respective masses of an atom of $He^3$ and of $He^4$, and X is the mole fraction of $He^3$.

Equation (1) is a solution to a boundary value problem which assumes that the characteristic boundary layer thickness is very small compared with the distance between the crystal surface and the electrode boundary wall. This assumption, of course, includes the requirements that the mean free paths be sufficiently small to provide shearing between "delta" layers of the liquid. The best theoretical estimates available of the mean free paths for this experiment (refs. 5 and 6) suggest that these conditions are met; however, for reasons to be discussed later in the section RESULTS AND DISCUSSION, the estimate for the mean free paths may be in error. For this reason, the viscosity obtained by using equation (1) in this experiment will be called the effective viscosity.

Part of the present results have been reported previously (ref. 10).

## EXPERIMENTAL PROCEDURE AND APPARATUS

A piezoelectric, cylindrical, quartz crystal was immersed in the liquid mixture and driven in a torsional mode of vibration at its resonant frequency of 11 kilohertz. The logarithmic decrements were then determined as a function of temperature by measuring the crystal resistance at resonance. This method for measuring the viscosity is a refinement of that first used by Welber and Quimby (refs. 11 and 12) and later by Webeler and Hammer (ref. 8).

A silver crystal holder served as a support for four pure silver electrode quadrants that surround, but do not contact, the crystal (see fig. 1). The crystal (0.53 cm in diam by 17.7 cm long) was supported by two nylon threads at its central strain node. The space between the electrode quadrants and the crystal surface (about 0.5 mm) was filled by the mixture of interest, and it is this portion of the total fluid (3/4 mole) in the copper sample chamber that provided the measured decrement.

The crystal resistance was measured by capacitance coupling to the electrode quadrants, which thereby eliminated the need for a mechanical contact. The electrode quadrants were connected to one arm of a modified Schering capacitance bridge by small electrical wires running inside a capillary tube from the sample chamber to room temperature. A PAR two-phase lock-in amplifier was used to detect the null bridge balance for very small driving voltages (0.1 to 0.3 mV) across the crystal at the crystal resonant frequency.

The low-temperature apparatus was a triple-reservoir system consisting of a $He^4$ bath at 1.0 K, a $He^3$ reservoir at 0.4 K, and a chromium-potassium alum (Cr-K alum) demagnetization cell (fig. 2). The demagnetization cell was thermally connected to the

sample chamber by means of slotted concentric copper cylinders. About 180 grams of Cr-K alum were packed between those portions of the concentric cylinders lying within the bore volume of the solenoidal magnet. To obtain good thermal contact between the salt and copper cylinders, the salt was first mixed with water and ground to the consistency of very fine silt and then mixed with a small amount of glycerine. This mixture was then packed with a special ramrod between the slotted concentric cylinders.

Two capillary tubes from the sample chamber pass through the Cr-K alum, $He^3$ reservoir, the $He^4$ bath, and finally extend to room temperature. One capillary tube carries electrical wires, and the other is used to fill the sample chamber with the sample fluid. All three reservoirs are necessary to avoid a large amount of heat transport into the sample chamber through the capillaries as a result of superfluid film flow and subsequent recondensation. In fact, when the $He^3$ reservoir temperature was increased to 0.7 K, the sample chamber warming rate increased more than 2 orders of magnitude.

The central tube (the function of which is subsequently described) is called the auxilliary heat exchange tube (see fig. 2). It passes through the $He^3$ reservoir and into the central core of the demagnetization cell. This tube connects the central core of the cell to a small tank of $He^3$ at room temperature. If a small amount of $He^3$ gas is condensed inside this tube, it can be used to reduce the temperature of the sample chamber and cell to nearly that of the $He^3$ reservoir before the working salt is demagnetized. This process is best illustrated by considering the expected sequence of steps during an experimental run. This sequence is begun by cooling all three reservoirs to 1.0 K, magnetizing the working salt to 17 kilogauss (1.7 T), and then evacuating the space inside the brass wall until a good vacuum is obtained. A small amount of $He^3$ gas is then condensed at 1.0 K inside the heat exchange tube, most of which ends up at the bottom of the cell core. A pure copper partition acts as the floor of the cell and cell core and and also as the ceiling of the sample chamber, thus providing thermal contact between these two compartments. This partition has a complicated shape and acts as a mechanical seal for the cell core, cell, and sample chamber. The $He^3$ reservoir, containing a substantial amount of liquid $He^3$, is now pumped to about 0.4 K. At this point in time, the sample chamber and demagnetization cell are still near 1.0 K. However, evaporation of the liquid $He^3$ droplets, located at the bottom of the central core of the demagnetization cell, followed by recondensation in the auxilliary heat exchange tube, where the tube passes through the $He^3$ reservoir, will cause the temperature of the sample chamber and cell to approach that of the $He^3$ reservoir (0.4 K). When the temperature of the sample chamber and cell are near 0.4 K, the Cr-K alum working salt is demagnetized over a 10- to 25-minute period. After the demagnetization is complete, the temperature of the sample chamber passes through a minimum value and thereafter rises.

The temperature of the liquid sample was determined from secondary thermometers. Four 10-ohm 1/10-watt Allen Bradley (carbon composition) thermometers (electrically in parallel) were used for temperature measurements near 1.0 K. Four 220-ohm, 1/2-

watt Speer carbon-composition thermometers (also in parallel) were used for temperature measurements below 1.0 K. These Speer resistors were lapped by use of a fine abrasive compound to fit into slotted holes in a pure copper block, which is thermally in contact with the top of the sample chamber. Two special low-resistance doped germanium thermometers (electrically in series) were also employed for temperature measurement below 1.0 K. These were placed in the bottom of the crystal holder. The germanium thermometers retain their calibration from one experiment to another and thus provide a check on any change in calibration of the more useful Speer thermometers (ref. 13). These secondary thermometers were calibrated by means of $He^3$ vapor pressure measurements at higher temperatures and by susceptibility measurements on a Cr-K alum pill at lower temperatures. The susceptibility measurements were performed in a separate experiment using a system similar to that described by Abel, Anderson, and Wheatley (ref. 14).

Measurements of the crystal decrement for $0.1 < T < 0.35$ K were made as the sample chamber and Cr-K alum cell warmed very slowly (about 10 mdeg/hr). The power dissipated by the crystal below 0.5 K was less than $2\times10^{-11}$ watt.

This same crystal and crystal holder has been used successfully to obtain viscosity values for pure liquid $He^4$ and pure $He^3$ (refs. 8, 15, and 16).

## RESULTS AND DISCUSSION

In figure 3 are shown the values of effective viscosity against temperature, as determined directly from the data, whereas table I shows smoothed values of the decrement $\Delta_l$ as a function of temperature. Data were obtained for 1.09-, 1.57-, and 4.67-percent molar solutions, hereinafter called 1.1S, 1.6S, and 4.7S, respectively. Two main facts are discernible from the data. As the temperature decreases, a slight increase in slope becomes noticeable near the Fermi temperature $T_f$. We calculated the $T_f$ values to be 0.13, 0.16, and 0.33 K, respectively, for 1.1S, 1.6S, and 4.7S. The experimentally identified $T_f$ for 4.7S is less than the calculated value of 0.33 K. This result is not surprising since the calculated $T_f$ values are for an ideal Fermi gas and these calculations become less applicable for higher concentrations of $He^3$. However, aside from Fermi-temperature-associated-slope changes, the solutions 1.1S and 1.6S also exhibit an even larger change in slope near 0.3 K, which is not near their Fermi temperatures. The viscosity for 1.1S and 1.6S continues to decrease down to the lowest measured temperature, while continuing to increase for 4.7S.

Although the authors cannot offer a reasonable explanation for the change of slope in $\eta_E$ for 1.1S and 1.6S, it is unlikely that changes in the mean free path are responsible. If any of the important mean free paths becomes comparable to the 0.5-millimeter

spacing between the crystal surface and the electrode quadrants, then the use of equation (1) to obtain values of $\eta_E \rho_{NS}$ would be invalid. The quasi-particle mean free path $\lambda_q$ may be estimated by

$$\lambda_q = v_q \tau_\eta$$

where $v_q$ is the velocity of a $He^3$ quasi-particle ($\sim 5 \times 10^3$ cm/sec) over the range of temperatures under consideration, and $\tau_\eta$ is the viscosity lifetime as computed in reference 6. Since $\tau_\eta$ never becomes larger than about $5 \times 10^{-8}$ second, $\lambda_q$ should be $<<0.5$ millimeter for all three mixtures throughout the entire temperature range.

The phonon mean free path is not as readily estimated. Baym and Ebner (ref. 5) have made theoretical estimates of the phonon mean free path for thermal conductivity that are comparable to the 0.5-millimeter value in question. The phonon mean free path for viscosity could well be of the same order of magnitude. However, this may be a moot point since the phonon population is so small at these temperatures ($T < 0.3$ K) that the contribution of the phonons to the crystal decrement should be much less than that due to the quasi-particles. For these reasons, the abrupt changes in the slope of $\eta_E$ for the 1.1- and 1.6-percent solutions do not appear to be mean free path effects.

A comparison of the present results with the theory of Baym and Saam (ref. 6) can be made by referring to figures 4 and 5, which show Baym and Saam's prediction for a 1.3- and 5-percent solution, respectively. Also appearing in these figures are the experimental results of Staas et al. (ref. 7) at temperatures above 1.0 K. The theoretical results appear to be high, although by less than would be guessed from Staas et al. The 5-percent solution can certainly be said to be in qualitative agreement with the theory. The situation in the case of the more dilute solutions is less clear cut. The aforementioned very sharp change in slope in the vicinity of 0.3 K is not predicted by the theory. Furthermore, there is a factor of 2 or more between the theoretical prediction and experimental values above 0.3 K, and the discrepancy increases below this temperature.

Figures 4 and 5 clearly indicate that the theoretical predictions are too high. Although the values obtained herein are consistently higher than those of Staas et al. (ref. 7) in the temperature region of 1 K in which there is an overlap of results, the theoretical results are still considerably higher. The theory is not applicable for $T > 0.6$ K. Even so, the extrapolation into this region should be smooth, and it seems clear that the theory overestimates the results, especially in the case of the dilute solutions. In this case, the disparity actually increases below 0.3 K, where the theory should be good. Perhaps the magnitude of the $He^3$ quasi-particle - $He^4$ phonon interaction is greater than the estimate used in arriving at the theoretical values. This possibility is mentioned indirectly by Baym and Saam, themselves, who suggest that the

theoretical $\tau_\eta$ may be too large.

Furthermore, the measurements of thermal conductivity made by Abel and Wheatley (ref. 17) show a disparity with the theoretical results of Baym and Ebner of about the same nature as our disagreement with Baym and Saam; that is, (1) there is an overall qualitative agreement, (2) the experimental results are consistently lower than the theoretical ones, (3) the results for the 1.3-percent solutions disagree more than the results for the 5-percent solutions, and (4) the largest discrepancy in the 1.3-percent results occurs in the region $0.1 < T < 0.6$ K. Abel and Wheatley also suggest that the discrepancy could result from a theoretical underestimate of the interaction between the $He^3$ quasi-particles and the $He^4$ phonons.

Although the sharp break in the data obtained herein at 0.3 K cannot be explained, it may be interesting to note that if, in figures 4 and 5, one makes the naive assumption that $\eta_3$ should be smaller by one-half, but $\eta_{ph}$ is correct, then the shape of $\eta_{total}$ reasonably approximates the experimental results.

Lewis Research Center,
National Aeronautics and Space Administration,
Cleveland, Ohio, September 29, 1969,
129-02.

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TABLE I. - SMOOTHED VALUES OF CRYSTAL DECREMENT DUE TO THE LIQUID AS FUNCTION OF TEMPERATURE FOR DILUTE SOLUTIONS OF HELIUM-3 IN SUPERFLUID HELIUM-4

| Temperature, K | Concentration, mole % | | | Temperature, K | Concentration, mole % | | |
|---|---|---|---|---|---|---|---|
| | 1.09 | 1.57 | 4.67 | | 1.09 | 1.57 | 4.67 |
| | Crystal decrement due to liquid, $\Delta_l$ | | | | Crystal decrement due to liquid, $\Delta_l$ | | |
| 0.10 | $0.910\times10^{-5}$ | $1.29\times10^{-5}$ | --------- | 0.36 | $1.29\times10^{-5}$ | $1.55\times10^{-5}$ | $2.71\times10^{-5}$ |
| .12 | .982 | 1.35 | --------- | .38 | 1.29 | 1.55 | 2.68 |
| .14 | 1.04 | 1.40 | --------- | .40 | 1.29 | 1.54 | 2.66 |
| .16 | 1.08 | 1.44 | $2.95\times10^{-5}$ | .45 | 1.29 | 1.52 | 2.60 |
| .18 | 1.11 | 1.465 | 2.92 | .50 | 1.285 | 1.495 | 2.54 |
| .20 | 1.145 | 1.485 | 2.90 | .60 | 1.275 | ----- | 2.47 |
| .22 | 1.17 | 1.50 | 2.88 | .70 | 1.28 | ----- | 2.42 |
| .24 | 1.20 | 1.52 | 2.85 | .80 | 1.295 | ----- | 2.40 |
| .26 | 1.23 | 1.54 | 2.83 | .90 | 1.32 | ----- | 2.38 |
| .28 | 1.25 | 1.55 | 2.805 | 1.00 | 1.365 | 1.54 | 2.38 |
| .30 | 1.27 | 1.56 | 2.78 | 1.10 | 1.49 | 1.63 | 2.39 |
| .32 | 1.285 | 1.565 | 2.76 | 1.12 | ---- | 1.77 | 2.43 |
| .34 | 1.29 | 1.56 | 2.73 | | | | |

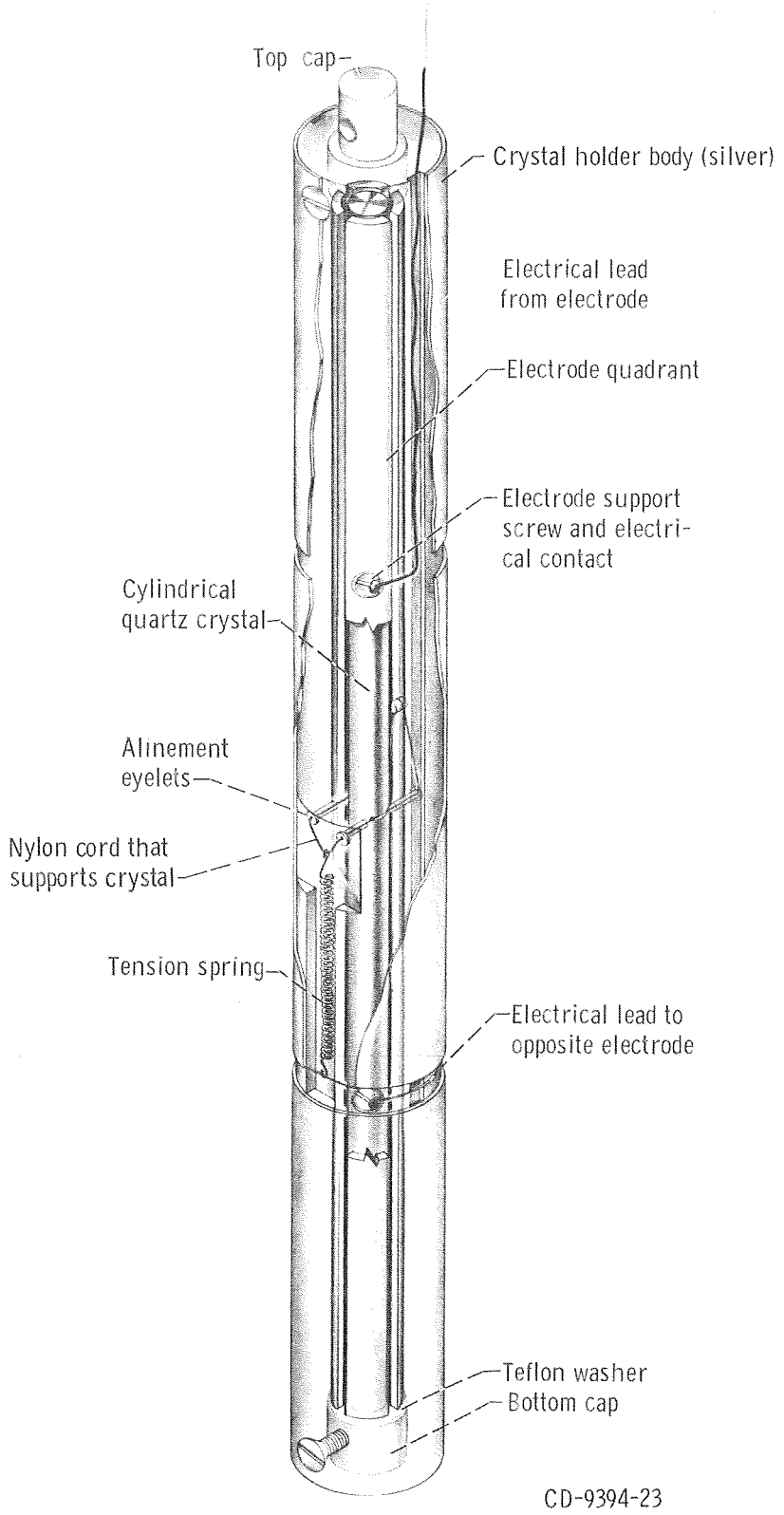


Figure 1. - Torsional crystal suspension system prior to placement in sample chamber of figure 2.

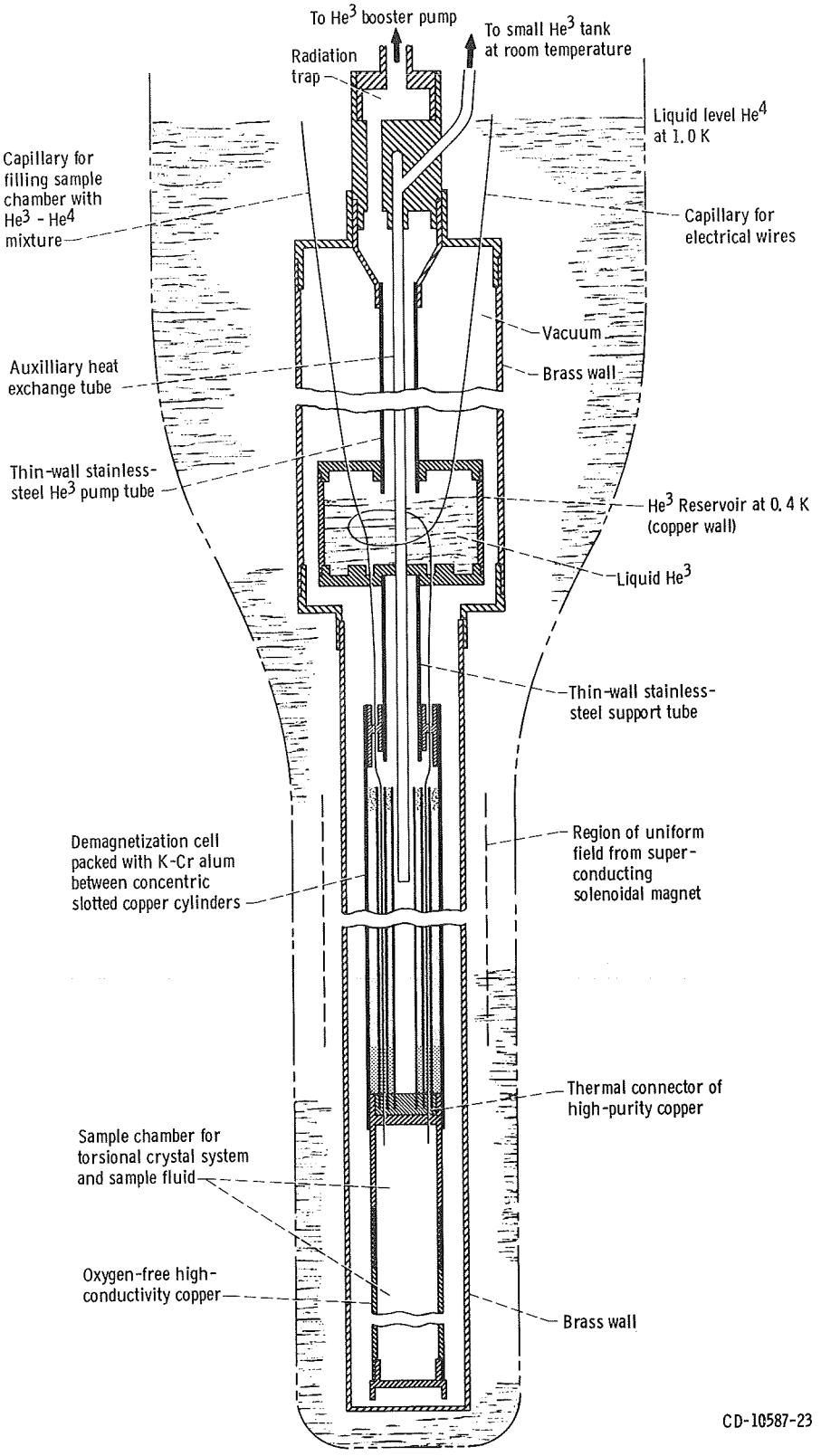


Figure 2. - Schematic of low-temperature apparatus showing only that part of triple-reservoir system located beneath surface of the 1.0 K $He^4$ bath.

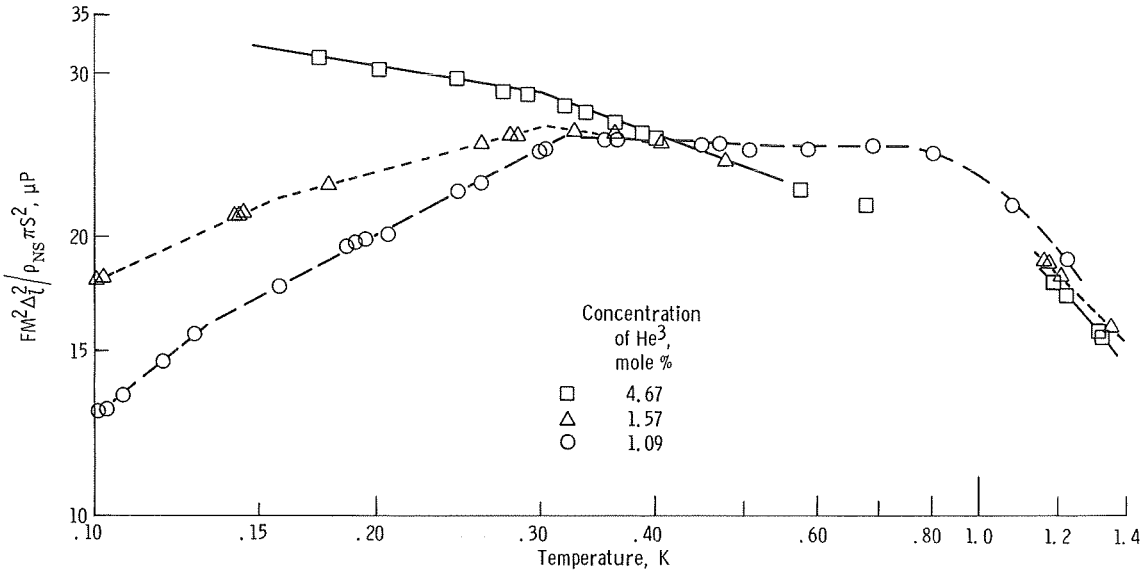


Figure 3. - Effective viscosity as function of temperature for dilute solutions of $He^3$ in superfluid $He^4$ (raw data). Mass of crystal, 10.477 grams; (surface area of crystal)$^2$, 898.7 centimeters$^4$; crystal resonant frequency, approximately 11 025 cycles per second; effective mass of $He^3$ atom, 2.34 $m_3$; maximum velocity of crystal rim, approximately 0.03 centimeter per second for temperatures less than 0.5 K.

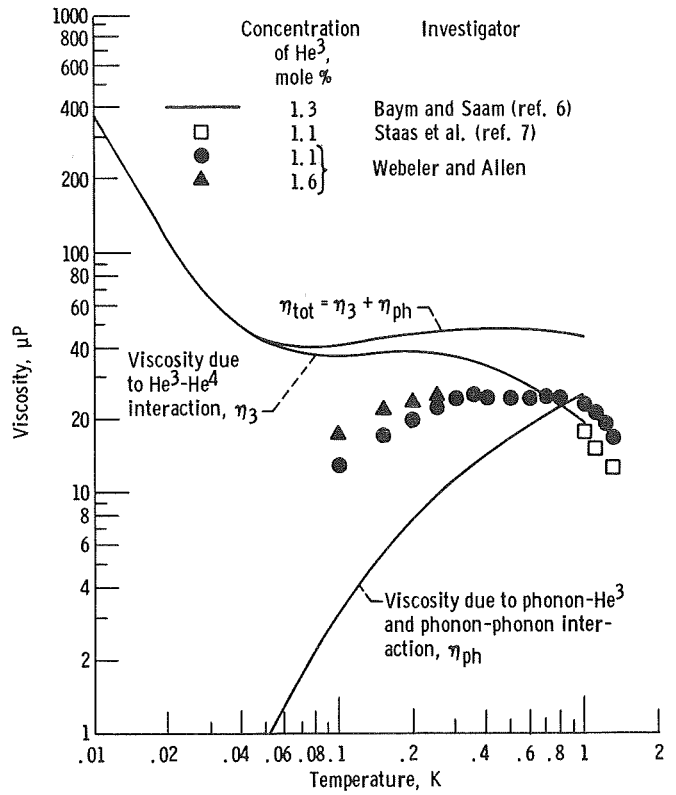


Figure 4. - Comparison of results obtained for viscosity as function of temperature for solutions of approximately 1.3 percent.

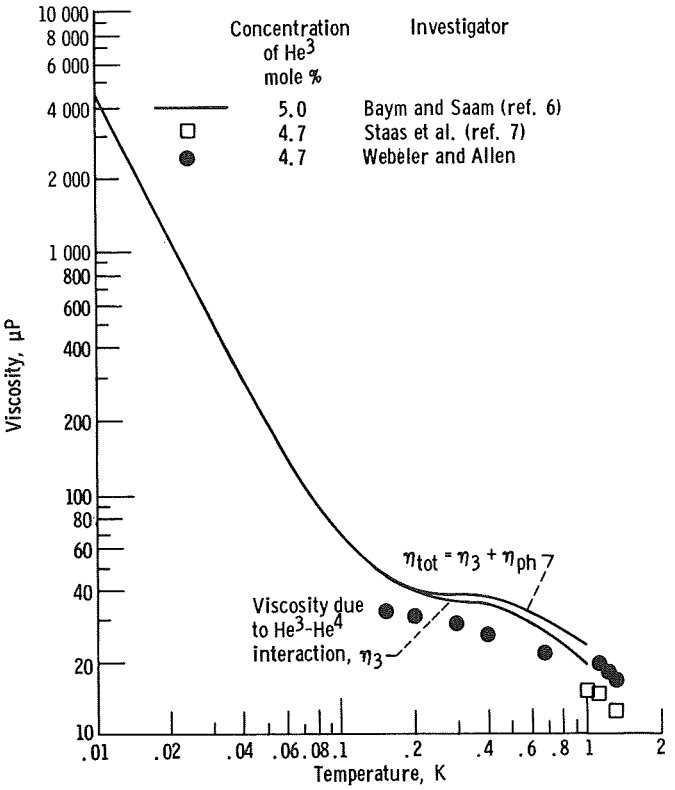


Figure 5. - Comparison of results obtained for viscosity as function of temperature for solutions of approximately 5 percent.

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